NATURAL RADIOACTIVITY, RADON CONCENTRATION AND HEAVY METALS IN SOIL AND WATER IN KEDAH, MALAYSIA

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NATURAL RADIOACTIVITY, RADON CONCENTRATION AND HEAVY METALS IN SOIL AND WATER IN KEDAH, MALAYSIA

By

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LIST OF ABBREVIATIONS

ADC	Analog-to- Digital Converter
CRM	Continuous Radon Monitor
DOE	Department of Environment
eV	Electron Volt
GPS	Global Positioning System
HPGe	High Purity Germanium
IAEA	International Atomic Energy Agency
ICRP	International Commission on Radiological Protection
ISO	International Organization for Standardization
NRPB	National Radiological Protection Board
NTDs	Nuclear Track Detectors
PCD	Pollution Control Department
RTC	Radon Tight Chamber
UNSCEAR	United Nations Scientific Committee on the Effects of Atomic Radiation
WHO	World Health Organization

LIST OF SYMBOLS

ε	Efficiency of the detector
η	Efficiency of the detector for the corresponding peak
λ_{Rn}	Decay constant of radon
$\boldsymbol{\lambda}$	Decay constant
ω	Back diffusion constant
A	Area of field of view
A_a	Surface area of sample
A_i	Intake of water
A_s	Specific Activity
A_{as}	Activity of the source
A_o	Initial activity
A_w	Radon in water
C_{eq}	Equilibrium radon concentration
C_f	Dose conversion factor
C_K	Activity concentrations of ^{40}K
C_{Ra}	Activity concentrations of ^{226}Ra
$C_{Rn}(t)$	Radon concentration measured by CRM
C_{Th}	Activity concentrations of ^{232}Th
D_c	Diameter of container
D_{in}	Indoor external dose
D_{out}	Outdoor external dose
D_s	Diameter of surface area of soil (used for CR-39)

E_d	Annual effective time
E_{indoor}	Indoor annual effective dose
$E_{outdoor}$	Outdoor annual effective dose
F_o	Radon Exhalation rate
H	Height of soil (used for CRM)
h	Height of container (used for CR-39)
H_{ex}	External hazard index
H_{in}	Internal hazard index
I_α	Alpha index
I_γ	Gamma index
L	Length of container (used for CRM)
n	Net area
N_i	No of tracks
P	Porosity of soil
$P_\gamma(E)$	Gamma ray emission probability at energy E
P_γ	Emission probability
S	Surface area of sample in RTC
T	Exposure time for CR-39 to measure radon in soil
t	Counting time
T_{eff}	Effective time for CR-39 to measure radon in soil
t_h	Decay time of the radionuclide
V	Volume of void space in container (used for RTC)
V_a	Volume of air in soil

V_{eff}	Effective volume of RTC
V_{equip}	Volume of equipments inside RTC
$V_{equip + soil}$	Volume of equipments and soil (CRM)
V_{soil}	Volume of container having soil (used for CRM)
V_{sw}	volume of soil and water
V_t	Volume of air dry soil
W	Width of container (used for CRM)
w	Weight of the sample
z_o	Soil thickness

LIST OF PUBLICATIONS

1. **Ahmad, N.,** Jaafar, M., & Alsaffar, M. (2015). Natural radioactivity in virgin and agricultural soil and its environmental implications in Sungai Petani, Kedah, Malaysia. *Pollution*, *1*(3), 305-313.
2. **Ahmad, N**., Jaafar, M. S., & Alsaffar, M. S. (2015). Study of radon concentration and toxic elements in drinking and irrigated water and its implications in Sungai Petani, Kedah, Malaysia. *Journal of Radiation Research and Applied Sciences*, 8(3), 265-276.
3. **Ahmad, N**., Jaafar, M. S., Bakhash, M., & Rahim, M. (2015). An overview on measurements of natural radioactivity in Malaysia. *Journal of radiation research and applied sciences*, *8*(1), 136-141.
4. **Ahmad, N**., Jaafar, M. S., & Khan, S. A. (2014). Correlation of radon exhalation rate with grain size of soil collected from Kedah, Malaysia. *Science International*, *26*(2).

KERADIOAKTIFAN TABII, KEPEKATAN RADON DAN LOGAM BERAT DALAM TANIH DAN AIR DI KEDAH, MALAYSIA

ABSTRAK

Radioaktif semula jadi, kepekatan ^{222}Rn dan logam berat (Pb, Ni, Cr, Cd dan As) telah ditentukan daripada 31 tanah tidak ditanam dengan sayuran, 42 tanah yang ditanam dan 51 air minuman dan air saliran untuk menyelidik kesan-kesan aktiviti manusia di Sungai Petani, Baling dan Kulim di Negeri Kedah, Malaysia. Kajian ini dijalankan dengan menggunakan Germanium berketulenan tinggi (HPGe) untuk mengukur keradioaktifan semula jadi, CR-39 NTDs dan Radon Monitor Berterusan (CRM) untuk mengukur kepekatan ^{222}Rn dalam tanah, Rad-7 untuk mengukur kepekatan ^{222}Rn dalam air dan Penyerapan Atom Spektrometer (AAS) untuk mengukur tahap logam berat dalam sampel tanah dan air. Kepekatan aktiviti purata ^{226}Ra, ^{232}Th dan ^{40}K didapati lebih tinggi di dalam tanah tanaman dan berada dalam lingkungan yang dilaporkan bagi negara-negara lain di seluruh dunia. Berdasarkan kepekatan aktiviti ^{226}Ra, ^{232}Th dan ^{40}K yang diselidik, indeks hazad luaran (seperti indeks gama (I_γ), aktiviti setara radium (Ra_{eq}), indeks hazad luaran (H_{ex}) dan dos luaran di luar ruangan (D_{out})), indeks hazad dalaman (seperti indeks alfa (I_α), indeks hazad dalaman (H_{in}) dan dos luaran di dalam ruangan (D_{in})) dan dos berkesan tahunan (seperti dos luar berkesan (E_{out}) dan dos dalaman berkesan (E_{in})) daripada sampel tanah telah dijumpai. Semua sampel tanah yang tidak digunakan dan digunakan untuk tanaman mempunyai aktiviti setara radium dalam tahap yang disyorkan, 370 Bq kg^{-1} yang dilaporkan oleh OECD, kecuali sampel dari Taman Desa Anggerik, Baling, Kampung

Stesen Guar, Kampung Kepala Bukit, Kampong Tandop, Kampung Dalam Wang, Kampong Janjung Merbau dan Kampung Bagan Sena. Kadar dos terserap luaran dan dalaman didapati lebih tinggi daripada had keselamatan 70 nGy h^{-1} dan 51 nGy h^{-1}, yang dilaporkan oleh UNSCEAR. Nilai purata H_{ex}, H_{in}, I_{α} dan I_{γ} dalam tanah tidak ditanam dan ditanam dengan sayuran didapati lebih rendah daripada satu, kecuali tanah tanaman di Baling, di mana nilai-nilai purata H_{in}, I_{γ} adalah sedikit tinggi daripada satu. Nilai-nilai dos berkesan tahunan dalaman dan dos berkesan tahunan luaran didapati di bawah had keselamatan 1 mSv y^{-1} untuk masyarakat awam yang disyorkan oleh ICRP. Keputusan yang diperoleh untuk kepekatan ^{222}Rn dalam tanah yang tidak digunakan untuk tanaman mempunyai saiz butiran berbeza menunjukkan kepekatan ^{222}Rn meningkat dengan peningkatan saiz butiran. Nilai kadar eskhalasi radon dari tanah tidak ditanam dan ditanam dengan sayuran didapati lebih rendah daripada had keselamatan 57.6 Bq m^{-2} h^{-1}. Nilai maksimum radon bawaan air didapati 20.0 ± 2.2 Bq/L di dalam air telaga dan minimum 1.4 ± 0.27 Bq/L dalam air paip. Nilai maksimum Pb, Ni, Cr, Cd dan As dalam tanah ditanam sayuran ditemui di ladang cili dengan nilai-nilai tertinggi masing-masing 2.29 ± 0.05 mg kg^{-1}, 2.76 ± 0.045 mg -kg^{-1}, 2.05 ± 0.029 mg/ kg, 0.52 ± 0.044 mg kg^{-1} dan 0.58 ± 0,042 mg kg^{-1}, dan minimum ditemui di ladang kelapa sawit dengan nilai terendah di bawah had pengesanan, 0.21 ± 0.022 mg kg^{-1}, di bawah had pengesanan, 0.03 ± 0.024 mg kg^{-1} dan 0.04 ± 0.006 mg kg^{-1}. Nilai maksimum Ni, Pb, Cd, Cr dan As dalam air ditemui di dalam air sungai dengan nilai-nilai tertinggi sebanyak 12.2 ± 1.2 μg/L, 9.74 ± 1.14 μg/L, 4.82 ± 0.72 μg/L, 5.4 ± 1.16 μg/L dan 7.2 ± 0.8 μg/L, dan nilai minimum ditemui dalam air paip dengan nilai terendah iaitu 0.28±0.1 μg/L bagi Ni dan 0.64 ± 0.14 μg/L, 0.1 ± 0.04 μg/L, 0.28 ± 0.06 μg/L dan di bawah had pengesanan bagi Pb, Cr, Cd dan As.

Walau bagaimanapun, semua sampel tanah dan air mempunyai kepekatan logam berat di bawah had keselamatan yang disyorkan oleh agensi yang berbeza.

NATURAL RADIOACTIVITY, RADON CONCENTRATION AND HEAVY METALS IN SOIL AND WATER IN KEDAH, MALAYSIA

ABSTRACT

Natural radioactivity, ^{222}Rn concentration and heavy metals (Pb, Ni, Cr, Cd and As) were determined in 31 uncultivated soil, 42 cultivated soil and 51 drinking and irrigated water to investigate the effects of human activities in Sungai Petani, Baling and Kulim in the state of Kedah, Malaysia. This study was conducted using High Purity Germanium (HPGe) to measure natural radioactivity, CR-39 NTDs and Continuous Radon Monitor (CRM) to measure ^{222}Rn concentration in soil, Rad-7 to measure ^{222}Rn concentration in water and Atomic Absorption Spectrometer (AAS) to measure the level of heavy metals in soil and water samples. The average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were found higher in cultivated soil and to be within those reported for other countries worldwide. Based on the investigated activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, outdoor hazard indices (such as gamma index (I_γ), radium equivalent activity (Ra_{eq}), external hazard index (H_{ex}) and outdoor external dose (D_{out})), indoor hazard indices (such as alpha index (I_α), internal hazard index (H_{in}) and indoor external dose (D_{in})) and annual effective doses (such as outdoor effective dose (E_{out}) and indoor effective dose (E_{in})) from soil samples were found. All the uncultivated and cultivated soil samples have radium equivalent activities within the recommended level 370 Bq kg^{-1} reported by OECD, except samples collected from Taman Desa Anggerik, Baling, Kampong Guar Station, Kampong Kepala Bukit, Kampong Tandop, Kampong Dalam Wang, Kampong Janjung Merbau and

Kampong Bagan Sena. Outdoor and indoor absorbed dose rates were found higher than the safety limits of 70 nGy h^{-1} and 51 nGy h^{-1}, respectively reported by UNSCEAR. The average values of H_{ex}, H_{in}, I_{α} and I_{γ} in uncultivated and cultivated soil were found lower than unity, except cultivated soil of Baling where the average values of H_{in}, I_{γ} were slightly higher than unity. The values of indoor annual effective dose and outdoor annual effective dose were found below the safety limit 1 mSv y^{-1} for general public recommended by ICRP. The results obtained for ^{222}Rn concentration in uncultivated soil having different grain size show that ^{222}Rn concentration increase with the increase in grain size. The values of radon exhalation rate from uncultivated and cultivated soil were found lower than safety limit 57.6 Bq m^{-2} h^{-1}. The maximum value of waterborne radon was found 20.0±2.2 Bq/L in well water and minimum was found 1.4±0.27 Bq/L in tap water. The maximum values of Pb, Ni, Cr, Cd and As in cultivated soil were found in chili farms with the highest values of 2.29±0.05 mg kg^{-1}, 2.76±0.045 mg kg^{-1}, 2.05±0.029 mg kg^{-1}, 0.52±0.044 mg/kg and 0.58±0.042 mg- kg^{-1}, respectively and minimum were found in palm oil farms with the lowest values of below detection limit, 0.21±0.022 mg kg^{-1}, below detection limit, 0.03±0.024 mg kg^{-1} and 0.04±0.006 mg kg^{-1}, respectively. The maximum values of Ni, Pb, Cd, Cr and As in water were found in stream water with the highest values of 12.2±1.2 µg/L, 9.74±1.14 µg/L, 4.82±0.72 µg/L, 5.4±1.16 µg/L and 7.2±0.8 µg/L, respectively and minimum were found in tap water with the lowest values of 0.28±0.1 µg/L for Ni and 0.64±0.14 µg/L, 0.1±0.04 µg/L, 0.28±0.06 µg/L and below detection limit for Pb, Cr, Cd and As, respectively. However, all the soil and water samples have heavy metals concentration below the safety limits recommended by different agencies.

CHAPTER 1

INTRODUCTION

1.1 Background

Humans are surrounded by radionuclides present in soil, air, water and human bodies. We ingest and inhale radionuclides on daily basis and radioactive materials have been ubiquitous on earth since it was formed. Radioactive materials found in nature are often referred to as Naturally Occurring Radioactive Materials, NORM (NCRP, 1987) and are categorized in three groups of radionuclides, namely primordial or terrestrial, cosmogenic and anthropogenic nature (UNSCEAR, 1988), which are everywhere in the environment. The primordial nuclides ^{238}U, ^{232}Th and ^{40}K are very long lived with half lives of 4.4×10^{9}, 1.4×10^{10} and 1.28×10^{9} years, respectively and are present since the earth was formed. These nuclides are produced by the process of nucleosynthesis in stars. The cosmogenic radionuclide's are continuously produced by the action of cosmic rays and are always present on the earth, even though they have half lives shorter than the life of the earth. More than 25 cosmogenic radionuclides have been identified. ^{14}C is typical example which is produced by the reactions $^{14}N(n,p)\ ^{14}C$ in atmosphere when the neutrons from cosmic rays interact with nitrogen (Lamarsh, 1983). The anthropogenic radioactivity is manmade radioisotopes (^{137}Cs, ^{131}I and ^{90}Sr) produced as a result of nuclear reactions with uranium. These nuclides are found everywhere as a result of nuclear weapons testing.

Fertilizers are usually used for cultivated purpose, which contain natural radio-activities like thorium, uranium and their decay product and traces of heavy metals (Olszewska-Wasiolek, 1995). Different types of fertilizers (containing phosphate) are used

to improve the growth of plants in the study area. Plants take some amount of radioactivity from the fertilizer applied to the soil. Humans are exposed externally and internally to radioactivity in rocks having phosphate and its by products. Gamma rays from phosphate rocks and fertilizers are the main sources of external exposure while radon, ingestion of fertilizer dust and radioactivity in food are the sources of internal exposure. Radionuclides of uranium and thorium decay series are relatively more abundant and naturally occurring. An example of the decay products of these series is the radon gas.

Radon is an odourless, tasteless and colorless gas, hence not easily detected. It is noble gas, due to which it is chemically inert. Radon is one of the heaviest noble gas at room temperature. It comprises three significant naturally occurring isotopes, ^{219}Rn, ^{220}Rn and ^{222}Rn. These isotopes belong to ^{235}U, ^{232}Th and ^{238}U decay series, respectively. The half lives of ^{220}Rn (Thoron), ^{219}Rn (Actinon), and ^{222}Rn (Radon) are 55.6 s, 3.96 s, and 3.83 days respectively.

^{222}Rn is the most significant among these isotopes because of its longer half life Therefore, this study focuses on ^{222}Rn. Other isotopes of radon are easily removed from atmosphere because of their short half lives.

For example, ^{219}Rn has approximately 0.7% abundance in the earth crust, which is attributable to its short half life and generally dissipates shortly after it is generated. Due to the short half life, ^{220}Rn decays before reaching the earth surface. The most significant isotope ^{222}Rn, can travel a considerable distance from its point of origin (Durrani & Ilic, 1997). That is why, only ^{222}Rn is regarded as a health hazard when estimating risk factors associated with radon exposure. Radon is an alpha emitter and considered as a foremost

source of lung cancer among non smokers and is the cause of 2900 deaths of non smokers worldwide (USEPA, 2004). Radon becomes airborne with the attachment of dust particle and pollution, after inhalation it becomes deeply trapped in the lungs, resulting in pathological effects like the decline in respiratory function (Khan et al., 2011). Beta particles are more hazardous than alpha particles due to longer penetration ability and are dangerous to skin. It has been investigated that emission of beta particle from strong sources burn the skin. In comparison to alpha and beta particles, gamma rays are most hazardous due to highest ability of penetration and are able to cross the body due which all organs of body could be effected (Alpen, 1997).

The term heavy metal is probably reserved for those elements with an atomic mass of 200 or greater (Baldwin & Marshall, 1999). It mostly comprises of some metalloids, transition metals, actinides and lanthanides (Appenroth, 2010). Commonly, the term has been used to any metal which is potentially toxic and/or clinically undesirable (Hardman, 2006). Most of heavy metals are toxic and their accumulation over time in the bodies of animals can cause severe diseases. Long-term exposure to heavy metals may result in progressing physical, neurological and muscular degenerative processes which may lead to Alzheimer's disease, muscular dystrophy and Parkinson's disease.

Lead is one of the most common toxic heavy metal while lead paint and lead water pipes are the major sources of the lead hazards. However, ore's smelting, battery manufacturing and traditional remedies are the second largest sources of lead poisoning (Baldwin & Marshall, 1999). Anthropogenic activities such as using of fertilizers, smelter emissions and sewage sludge to land are the most important sources of cadmium release to natural environment (Hutton & Symon, 1986). Industrial effluents and airborne particles

from combustion of fossil fuels are the main causes of nickel contamination of hydrosphere and atmosphere. Chromate is the common ore of chromium, commonly used to manufacture, amongst other things, cement, paints, leather products and anti-corrosives which directly contaminate the environment (Pradhan, 2012).

Measurement of natural radioactivity is of interest worldwide. A very limited data about the natural radioactivity in soil is available for Kedah. Almayahi et al. (2012b) found natural radioactivity in soil of Kedah with the maximum values of 79 Bq kg^{-1} for ^{226}Ra, 97 Bq kg^{-1} for ^{232}Th and 602 Bq kg^{-1} for ^{40}K and minimum values of 33 for ^{226}Ra, 81 Bq kg^{-1} for ^{232}Th and 270 Bq kg^{-1} for ^{40}K.

1.2 Problem Statements

Human beings depend on soils and good soils depend on human beings and the use they make on them. Soil exists as a mixture of naturally occurring materials on the surface of earth having supporting plants and living bodies. Human activities such as using of fertilizers in improving the properties of plants and reclaiming the land and industrialization can change the soil concentrations. Using fertilizers for long term could enhance the concentrations of natural radioactivity and heavy metals and consequently increase the radiological hazards which would increase the diseases for human beings (El-Farrash et al., 2012). Human beings are exposed to natural radioactivity, radon and heavy metals by consuming contaminated water, plants and animals which result in various biochemical disorders. Sungai Petani, Baling and Kulim have agricultural activity more than other areas of Kedah and have industrial area. Different types of fertilizers are used for improving the properties of plants like Chili, Banana and Palm Oil in the studied areas.

Therefore, the knowledge of the distribution and concentrations of natural radioactivity, radon concentration and heavy metals are of interest since it gives very important information in the monitoring of environmental contamination. This research interests in investigating the following problems.

a. What is the level of natural radioactivity in uncultivated soil and cultivated soil from chili, banana and palm oil farms?
b. What is the level of radon concentration in soil, drinking and irrigated water?
c. What is the correlation of radon concentration with grain size of soil? What is the effect of grain size of soil with radon exhalation rate?
d. To find the concentrations of heavy metals (Ni, Cd, As, Pb, Cr) in cultivated soil from chili, banana and palm oil farms and water?

1.3 Objectives of the Research

The objectives of this research are:

1. To measure the concentrations of natural radioactivity in uncultivated soil and cultivated soil from chili, banana and palm oil farms.
2. To determine the Rn-222 concentration in soil, drinking and irrigated water.
3. To find the correlation between radon concentration and grain size of soil.
4. To find the concentrations of heavy metals (Ni, Cd, As, Pb, Cr) in cultivated soil from chili, banana and palm oil farms and water.

1.4 Scope of Research

This study was focused on measurements of natural radioactivity and radon concentration in uncultivated and cultivated soil collected from palm oil, chili and banana farms and on radon concentration in water used for drinking and irrigation in Kedah, Malaysia. Unfortunately a very limited data are available in literature for radon concentration in soil and water. This study is important as it provides a baseline data for natural radioactivity, radon concentrations and heavy metals concentrations in cultivated soil and irrigated water. These were measured by High Purity Germanium (HPGe), CR-39, Continuous Radon Monitor (CRM), RAD-7 and Atomic Absorption Spectrometer (AAS).

1.5 Outline of Thesis

This thesis includes five chapters. Chapter 1 is the background of natural radioactivity, problem statements, and objectives of the research and scope of the research. Chapter 2 summarized the natural radioactivity, radon concentration as well as literature review on natural radioactivity, radon concentration and heavy metals. Chapter 3 provides descriptions of the study area, samples collection and materials and methods whilst Chapter 4 provides the results and discussion. Finally, Chapter 5 presents conclusion and future work related to this research.

CHAPTER 2

THEORY

2.1 Environmental Natural Radioactivity

Uranium and thorium naturally occurs randomly, although in small quantities all over the earth's crust, typically at ppm levels. However, there are specific places where the concentration is high (UNSCEAR, 1988). ^{238}U and ^{232}Th are naturally occurring radionuclide's and are the parent elements of the two radioactive decay series. Their decay products are alpha, beta and gamma rays emitters. Uranium occurs naturally in the form of ^{234}U, ^{235}U and ^{238}U. The relative abundance of ^{238}U is 99.274% and the equilibrium concentration of ^{234}U is 0.0054%. The relative abundance of ^{235}U is 0.7205%. ^{234}U is a member of ^{238}U decay series. The contribution of ^{235}U in the natural pollution is negligible because of its relatively low abundance (IAEA, 1990). ^{238}U and ^{235}U decay series are shown in Fig 2.1 and 2.2, respectively.

The ^{238}U series has fifteen members' ends up to ^{206}Pb after 8 alpha and 6 beta emissions along with many gamma decays. Typical concentration of uranium in Granite, Gabbro, Limestone and Sandstone is 3 to 5, < 1, 1 to 2 and 3 to 5 ppm, respectively with average value of 2.7 ppm. Natural thorium consists almost entirely of ^{232}Th, 1.35×10^{-8} % of ^{228}Th and extremely small amount of ^{234}Th, ^{230}Th, ^{231}Th and ^{227}Th. ^{232}Th is the parent of 4n (n varying from 58 to 51) radioactive decay series (Kaplan, 1972). There twelve members in the series and ^{232}Th, as shown in Fig 2.3, after 7 alpha and 5 beta emissions along with many gamma radiations decays in to ^{208}Pb. The range of concentration on ^{232}Th on the earth's crust varies from zero to several hundreds of parts per million (ppm).

Typical concentration of thorium in Granite, Gabbro, Limestone and Sandstone is 10 to 30, 2 to 3, 1 to 2 and 10 to 15 ppm, respectively with average value of 9.6 ppm (IAEA, 1990). Among the naturally occurring potassium isotopes, ^{40}K is unstable. It has a half life of 1.227×10^{9} years.

The relative abundance of ^{40}K in natural potassium is 1.18×10^{-4}. It decays by $^{-}\beta$ –decay to ^{40}Ca and by $^{+}\beta$-decay or electron capture (K-capture) to ^{40}A. The composition of potassium (K) in rocks ranges from 0 to 10 %, typically 1 to 5 % with a mean value of 2% (IAEA, 1990). A similarly significant daughter from of the uranium decay series existing in the environment is ^{226}Ra, which is the likely emitter of natural radioisotopes ^{222}Rn, the radon gas. Human exposure to high concentration of radon and its progenies for lengthy period result in the decline of respiratory functions and emergence of lung cancer (Verma et al., 2012). Radon and its decay product have more than 50% contribution to the total effective dose. (UNSCEAR, 2000b). Thus, radon and its decay products have garnered a lot of interest because of their health hazards, as these radionuclides may attain fairly dangerous levels in dwelling with the lack of sufficient ventilation system or contain strong sources of radon. Therefore, measurement of radon are being performed worldwide at national levels to generate extensive data, which are openly accessible (Almayahi et al., 2011; Faheem, 2008; Ismail & Jaafar, 2013; Rahman, 2006; Saad et al., 2013; Singh et al., 2010; Verma et al., 2012).

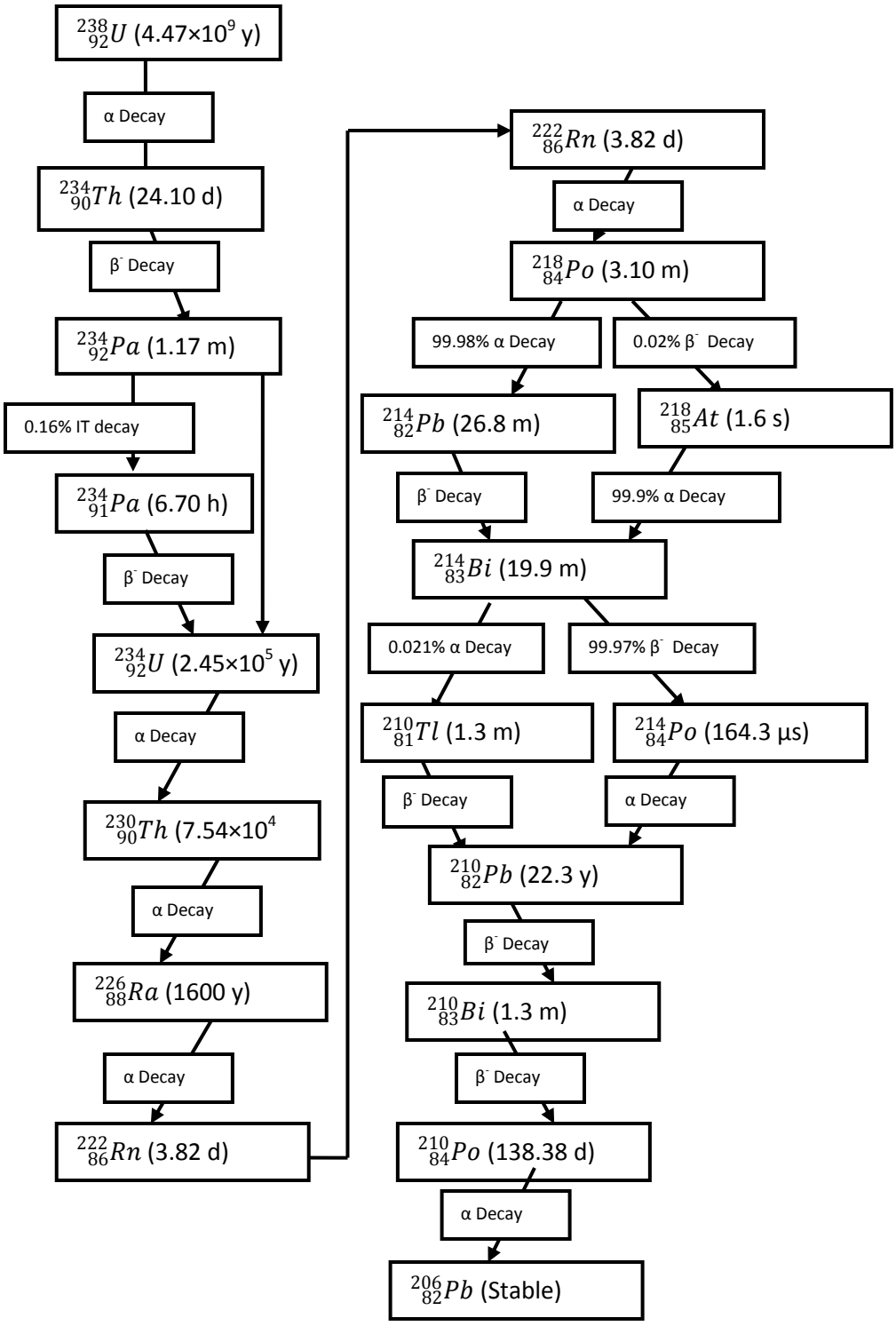

Figure 2.1: Uranium-238 decay series (Malain, 2011)

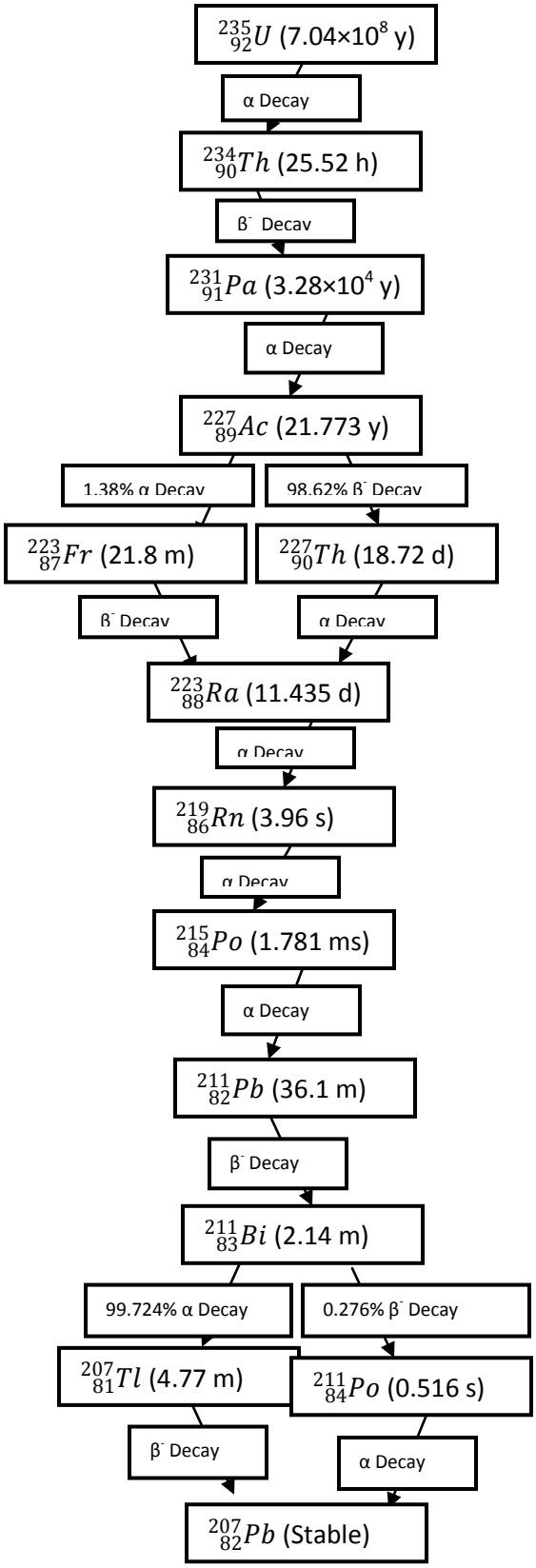

Fig 2.2: Uranium-235 decay series (Malain, 2011)

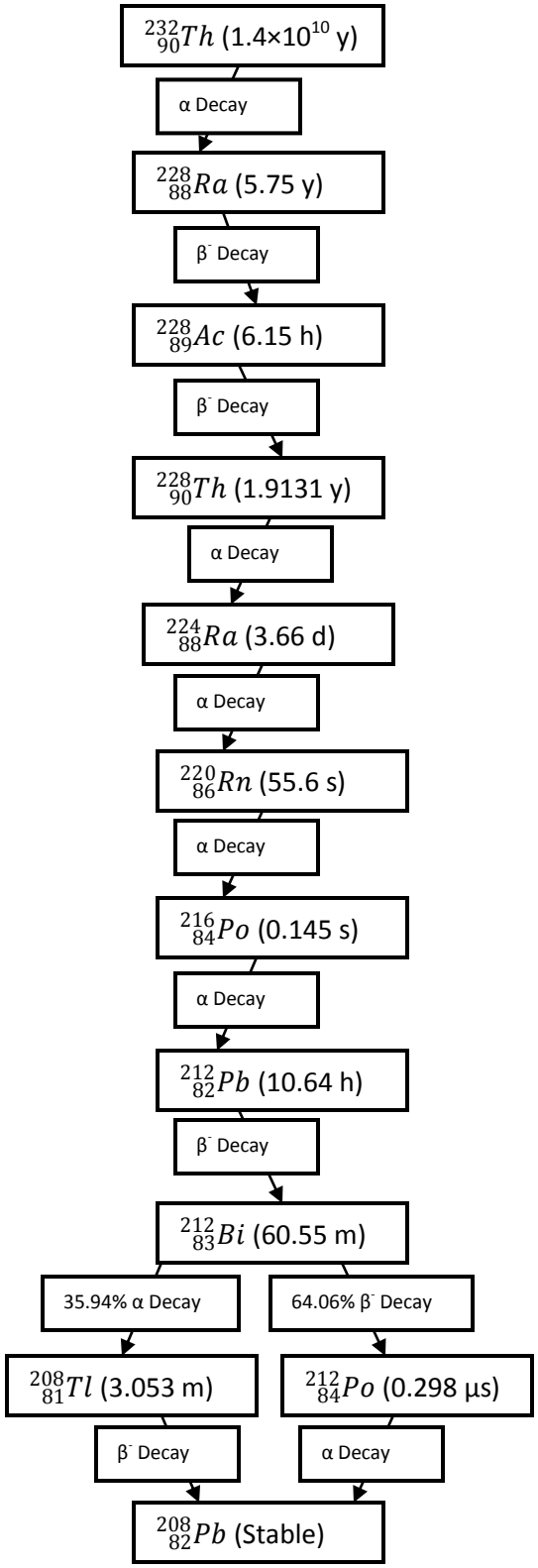

Fig 2.3: Thorium-232 decay series (Malain, 2011)

2.2 Radon Emanation

Radon occurs in nature due to the decay of radium in mineral grain. Most of the radon produced continues to adhere to the grain particles, while a small fraction permeates into the pore spaces either rapidly or within a few days before it eventually decays (Duenas et al., 1997). The ratio of the radon released from the grain to the produced radon in the grain is measured as the co-efficient of emanation (E).

The quantity of emanated radon to pore spaces is dependent on the spatial distribution of ^{226}Ra contained in the mineral grain, the radium concentration and pore moisture content (Sasaki et al., 2004). Huge amounts of radon concentrations results generally from minor disparity in radium concentration in the soil. This variation in radon concentration is attributable to random distribution of radium in grains.

2.3 Radon Exhalation

The movement of radon from source environment such as construction sites, building materials and soil to indoors is referred to as radon exhalation. Exhalation rate is the amount of atoms escaping the soil per unit surface area per unit time. It is used to measure exhalation. The exhalation rate of radon is determined to a large extent by atmospheric pressure, forces of wind and temperature. A large volume of small pores are filled with water under such conditions resulting in high exhalation rate (Sun et al., 2004).

2.4 Transport of Radon

Most radon produced by the decay of radium never escapes from its birth mineral; instead it is usually lodged firmly in position inside the crystal lattice for few days pending

its decay. The minute fraction of radon that escapes is either released quickly as soon as it is born or within the few days prior to it decays.

The first option for escape is the direct ejection of the radon atom by recoil from alpha emission (Kigoshi, 1971). In relation to the conservation of momentum, the emission of an alpha particle with 4.78 MeV by ^{226}Ra provides the remaining ^{222}Rn nucleus recoil energy of 86 keV, which is enough to initiate the recoil motion of radon through 26 nm of SiO_2. If the radium exists at a distance of 26 nm from the surface of the mineral, the recoil can really dislodge from the grain and go into interstitial space.

If the pore space filled with water, the dislodged recoil most likely ejects into the liquid as illustrated as shown in Figure 2.4. The radon atom subsequently diffuses from the water or be moved by it.

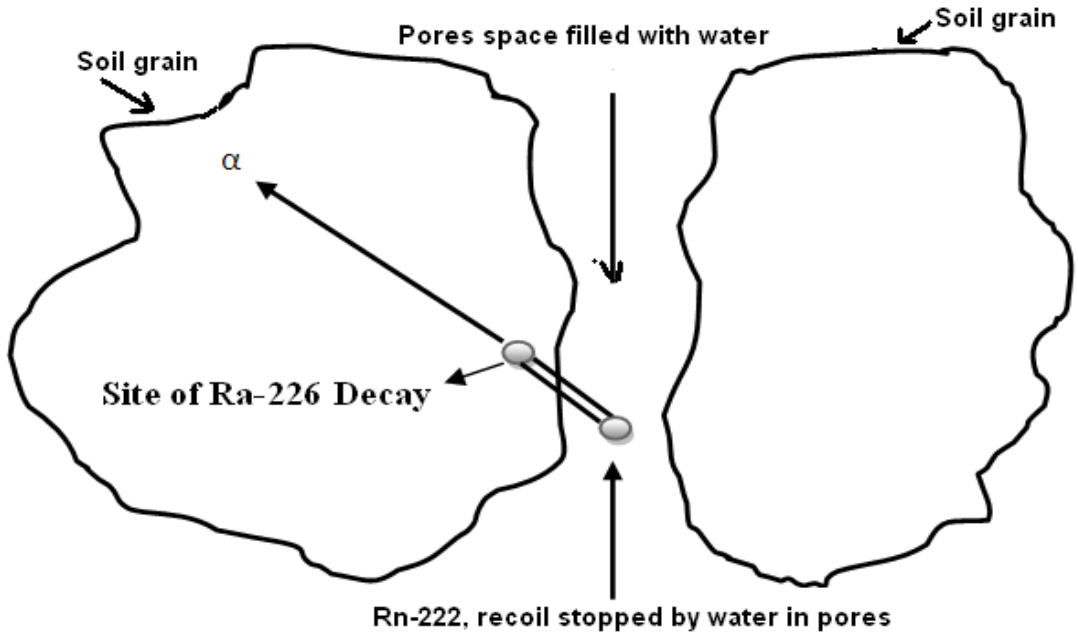

Figure 2.4: Rn-222 stopped by water in pores

The second possibility depicted in Figure 2.5 is for a case where the interstitial space is dry (filled only with soil gas) and not sufficiently wide to impede the recoiling radon. Thus the recoil is ejected to an adjacent grain. If the initially dry grains become wet prior to radon decay, it can be discharged into the interstitial space, from where it can be diffused (Fleischer, 1980).

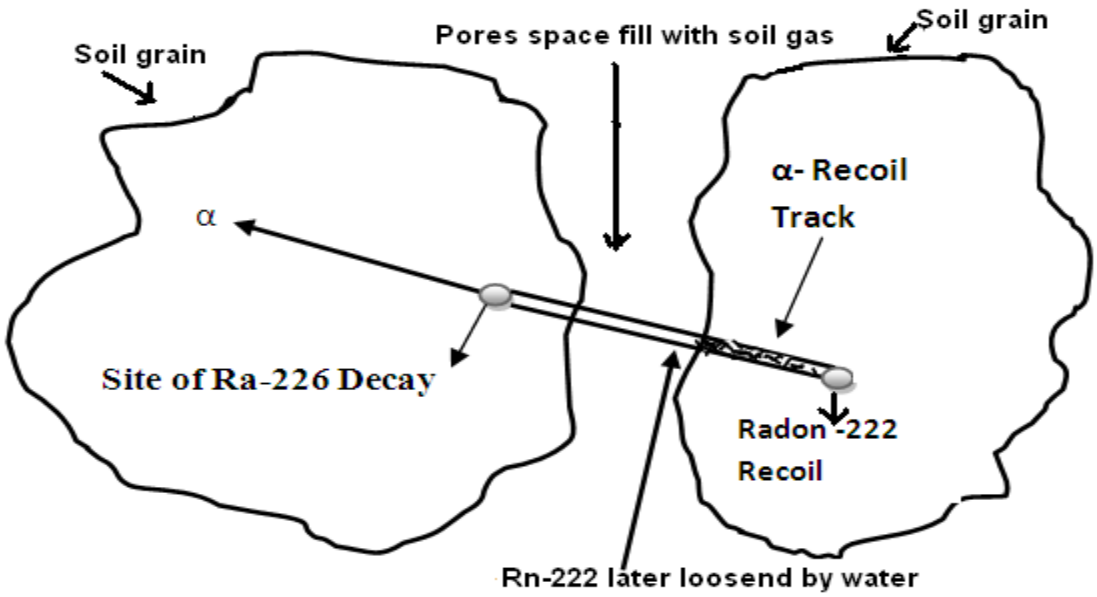

Figure 2.5: Rn-222 later loosened by water

2.5 Literature Review

Numerous studies have conducted throughout the world to find the natural radioactivity, radon concentration, and heavy metals in soil, water by using different methods. Natural radioactivity in soil, radon concentration in soil and water and heavy metals in water measured by different researchers worldwide are summarized in Table 2.1, 2.2, 2.3 and 2.4, respectively.

Table 2.1: Measurements of natural radioactivity in soil worldwide

Sample location	Sample type	Method	Results	References
Palong, Johor, Malaysia	Soil	Neutron Activation Analysis (NAA)	^{238}U= 58.8- 484.8 Bq/kg ^{232}Th =59.6-1204 Bq/kg. The concentrations of ^{238}U and ^{232}Th were found higher in all except two locations (sample S2 and S5)	(Ramli et al., 2005)
Ulu Tiram, Malaysia	Soil	NaI gamma ray detector	^{238}U= 1.74 - 4.58 ppm (mean: 3.63 ppm) ^{232}Th=(10.68- 82.10 ppm) (mean: 43.00ppm)	(Abdul Rahman & Ramli, 2007)
Kinta, Malaysia	Soil	High Purity Germanium detector (HPGe)	^{238}U=12 – 426 Bq kg^{-1} ^{232}Th =19 -1377 Bq kg^{-1} ^{40}K = 19 - 220 Bq kg^{-1} External gamma dose rate = 222 nGy h^{-1}	(Lee et al., 2009)
Research Station, Nation Park, Malaysia	Soil	High Purity Germanium detector (HPGe) and Portable Radiation Survey Meter	^{40}K= 598.24 Bq kg^{-1} ^{226}Ra = 99.13 Bq kg^{-1} ^{228}Ra= 139.98 Bq kg^{-1} Mean of doses were 0.215µSv/h and 0.193 µSv/h on the ground and one meter from ground, respectively.	(Saat et al., 2011)

Table 2.1 continued

Penang, Malaysia	Soil	High Purity Germanium detector (HPGe)	^{40}K = (mean: 835 Bq kg^{-1}) ^{226}Ra = (mean: 396 Bq kg^{-1}) ^{238}U = (mean: 184 Bq kg^{-1}) ^{232}Th = (mean: 165 Bq kg^{-1}) The values of radium equivalent activity (Ra_{eq}), external (H_{ex}) and internal hazard indices (H_{in}), annual gonadal dose equivalent, absorbed dose rates in indoor air, effective dose equivalent rate and $^{226}Ra/^{238}U$ were found 696 Bq kg^{-1}, 1.87, 2.9, 2.02 mSv y^{-1}, 315 nGy h^{-1}, 0.38 mSv/y and 2.10, respectively.	(Almayahi et al., 2012a)
Northern Peninsular, Malaysia	Soil	High Purity Germanium detector (HPGe)	^{40}K = (mean: 427 Bq kg^{-1}) ^{226}Ra = (mean: 57 Bq kg^{-1}) ^{232}Th = (mean: 68 Bq kg^{-1}) The mean values of $Ra_{eq,}$ H_{ex} and H_{in} were found as 186 Bq kg^{-1} , 0.50 and 0.65, respectively while that of annual effective dose rates (ED) and absorbed dose rates (D_R) were found, 108 µSv y^{-1} and 88 nGy h^{-1}, respectively. Health hazard indices were found higher (1.1 H_{ex}) and (1.1 H_{ex}, 1.6 H_{in}) only in two samples.	(Almayahi et al., 2012b)
Perak, Malaysia	Soil	High Purity Germanium detector (HPGe)	^{238}U = (mean: 127 Bq kg^{-1}) ^{232}Th = (mean: 304 Bq kg^{-1}) ^{40}K = (mean: 302 Bq kg^{-1}) External hazard index (H_{ex})= 0.35-3.07	(Heru Apriantoro & Termizi Ramli, 2013)

Table 2.1 continued

Jordan	Soil	High Purity Germanium detector (HPGe)	^{226}Ra = (range: 43.2-228.9 Bq kg^{-1}) ^{232}Th = (range: 17.9-31.9 Bq kg^{-1}) ^{40}K = (range: 290.0-558.4 Bq kg^{-1}) Average radium equivalent activities were found within acceptable limits.	(Ahmad & Khatibeh, 1997)
Jordan	Soil	High Purity Germanium detector (HPGe)	^{238}U = (range: 22-104Bq kg^{-1}) ^{232}Th = (range: 21-103 Bq kg^{-1}) ^{40}K = (range: 138-601 Bq kg^{-1})	(Al-Jundi et al., 2003)
Malwa, Punjab, India	Soil	High Purity Germanium detector (HPGe)	^{226}Ra = (range: 18.3-53.1 Bq kg^{-1}) ^{232}Th = (range: 57.2-148.2 Bq kg^{-1}) ^{40}K = (range: 211.1-413.2 Bq kg^{-1}) The values of dose rate (D_R) ranged from 58.08 to 130.85 nGy h^{-1} with an average of 79.11 nGy h^{-1}. The values of external hazard index ranged from 0.35 to 0.79.	(Mehra et al., 2007)
South Konkan, India	Soil	High Purity Germanium detector (HPGe)	^{238}U = (mean: 44.97 Bq kg^{-1}) ^{232}Th = (mean: 59.70 Bq kg^{-1}) ^{40}K = (mean: 217.51 Bq kg^{-1}) Average absorbed dose rate was found 68.08 nGy h^{-1}. Radium equivalent activity was found below the recommended value.	(Dhawal et al., 2013)
Punjab, Pakistan	Soil	High Purity Germanium detector (HPGe)	^{226}Ra = (range: 20-43 Bq kg^{-1}) ^{232}Th = (range: 29-53 Bq kg^{-1}) ^{40}K = (range: 98-621 Bq kg^{-1}) The estimated values of ED, H_{in}, H_{ex} and Ra_{eq} were found within recommended values.	(Faheem & Mujahid, 2008)

Table 2.1 continued

Azad Kashmir, Pakistan	Soil	High Purity Germanium detector (HPGe)	^{226}Ra = (range: 10-47 Bq kg^{-1}) ^{232}Th = (range: 18-75 Bq kg^{-1}) ^{40}K = (range: 40-683 Bq kg^{-1}) The reported values of radium equivalent activity, annual effective dose and hazard indices were found within acceptable limits.	(Rafique et al., 2011a)

Table 2.2: Measurements of radon concentration in soil worldwide

Sample location	Sample type	Method	Results	References
Pakistan	Soil, sand	CR-39 NTDs	Radon exhalation rate in soil samples collected from Bahawalpur Division and NWFP ranged from 1.56 to 3.33 Bq m^{-2} h^{-1} and 2.49 to 4.66 Bq m^{-2} h^{-1}, respectively. In case of sand samples its values ranged from 2.78 to 20.8 Bq m^{-2} h^{-1} and 0.99 to 4.2 Bq m^{-2} h^{-1}, respectively.	(Rahman, 2006)
NW Slovenia	Soil	Alpha Guard Radon Monitor	Values of radon concentrations ranged from 0.9 to 32.9 kBq m^{-3}, while radon exhalation rate ranged from 1.1 to 41.9 mBq m^{-2} s^{-1}.	(Vaupotič et al., 2010)
North Malaysia	Soil	CR-39 NTDs	The maximum radon concentration was found 375.42 kBq m^{-3} and minimum was found 2.23 kBq m^{-3}.	(Almayahi et al., 2011)
Malaysia	Fertilizer	CR-39 NTDs	The radon concentration ranged from 79.25 ± 23.24 to 634.01 ± 51.42 Bqm^{-3}.	(Aswood et al., 2014)
Malaysia	Soil	CR-39 NTDs	Radon concentration ranged: 2,225 to 9,950 Bq m^{-3}	(Almayahi et al., 2014)

Table 2.3: Measurements of radon concentration in water samples worldwide

Sample location	Sample type	Method	Results	References
Karnatak, India	Ground water	RAD-7	The radon concentrations in Varahi command area ranged from 0.2±0.4 to 10.1±1.7 Bq L^{-1} having average value of 2.07±0.84 Bq L^{-1}, while in case of water samples collected from Markandeya command area its values ranged from 2.21± 1.22 to 27.3±0.787 Bq L^{-1} having average value of 9.30±1.45 Bq L^{-1}	(Somashekar & Ravikumar, 2010)
Islamabad and Murree, Pakistan	Water	RAD-7	The radon concentrations in water and soil samples from Islamabad region ranged from 25.90 to 158.40 kBq m^{-3} and 17.34 to 72.52 kBq m^{-3} with the mean values of 88.63 kBq m^{-3} and 45.08 kBq m^{-3}, respectively. In Murree and its surroundings its values ranged from 1.64 to 10.20 kBq m^{-3} and 0.61 to 3.89 kBq m^{-3} having mean values of 4.38 kBq m^{-3} and 1.70 kBq m^{-3}, respectively.	(Ali et al., 2010)
Punjab, India	Ground water	RAD-7	The concentrations of radon ranged from 2560 to 7750 Bq m^{-3} with an average value of 5143.33 Bq m^{-3}. The absorbed dose rate ranged from 1.26 to 3.24 mSv y^{-1}.	(Badhan et al., 2010)
Iraq	Water	RAD, CR-39 NTDs	Minimum value of radon concentration was found 174 Bq m^{-3} in Tap water, while maximum was found 2050 Bq m^{-3} in well water. In case of oil-production water its values ranged from 8464 to 5092 Bq m^{-3}.	(Subber et al., 2011)

Table 2.3 continued

Penang, Malaysia	Water	RAD-7	The estimated radon concentrations ranged from 0.49 to 9.72 Bq L^{-1}, 0.58 to 2.54 Bq L^{-1} and 7.49 to 26.25 Bq L^{-1} in treated, bottled and raw water, respectively. The committed effective doses from radon were estimated were ranged from 0.003 to 0.048 mSv y^{-1}, 0.001 to 0.018 mSv y^{-1} and 0.002 to 0.023 mSv y^{-1}, for 0 to 1, 2 to16 and > 16 y age groups, respectively.	(Muhammad et al., 2012)
Cameron Highlands, Malaysia	Irrigation water	RAD-7	Average radon concentrations were ranged from 0.21 to 0.297 Bq L^{-1}.	(Al-Nafiey et al., 2014)

Table 2.4: Measurements of heavy metals in water samples worldwide

Sample location	Sample type	Method	Results	References
Southwestern Turkey	Stream water	ICP-AES	The mean values of Cd, Cu, Pb, Zn and Cr were found 0.800 ± 0.600 µg/L, 13.000 ± 9.000 µg/L, 83.600 ± 56.200 µg/L, 37.000 ± 26.000 µg/L and 19.700 ± 15.600 µg/L, respectively.	(Demirak et al., 2006)
Egypt	Lakes water	Atomic Absorption Spectrometer	The concentrations of Fe, Zn, Cu, Mn, Cd and Pb were found 1.42 mg/L, 0.4636 mg/L, 0.513 mg/L, 0.513 mg/L, 0.044 mg/L and 0.099 mg/L, respectively. The order of concentrations were found Fe > Mn > Pb > Zn > Cu > Cd in Lake Edku, whereas Fe > Mn > Pb > Zn > Cu > Cd in Lake Borollus. Its order was found Fe > Mn = Cu > Zn > Pb > Cd in Lake Manzala.	(Saeed & Shaker, 2008)
China	Sea water	Atomic Absorption Spectrometer	The concentrations of Zn, As, Pb, Cd and Cu were ranged from 2.4 to 52.4µg/L, 1.41 to 2.98 µg/L, 0.35 to 1.70 µg/L, 0.04 to 1.0 µg/L and 0.03 to 1.18 µg/L for Zn, As, Pb, Cd and Cu, respectively.	(Wang et al., 2010)
Malaysia	Tap water	Atomic Absorption Spectrometer	The mean concentrations of heavy metals (Ni, As, Cd and Pb) were found 0.91 µg/L, 0.81 µg/L, 0.41 µg/L and 0.28 µg/L.	(He et al., 2011)

CHAPTER 3

MATERIALS AND METHODS

3.1 Area under Study

This study was conducted in selected locations of Kedah. Kedah is a state of Malaysia, situated in the north part of Peninsular Malaysia and covers an area of 9,427 km^2 (3,640 square miles). It is located at 6º 07´ 42´´ N 100º 21´ 46´´ E on the world map. The north part of Kedah borders the state of Perlis and shares an international boundary with Thailand. In south and southwest it borders the states of Perak and Penang, respectively. Kedah has tropical climate having uniform temperature and average humidity ranged from 82% to 86% per annum. Average annual rain fall ranged from 203 cm to 254 cm. Geologically it is divided into the following groups: Silurian-Ordovician, Triassic, Quaternary, Cretaceous-Jurassic, Carboniferous and Cambrian as shown in Figure 3.1. Samples were collected from Sungai Petani, Kulim and Baling. The locations of cultivated areas were selected according to the suggestions of cultivated departments of Sungai Petani, Kulim and Baling as these locations were registered with cultivated departments.

Sungai Petani is a capital of district Kuala Muda in the state of Kedah, and covers an area of 925 km^2. It is located at 5º 38´ 49´´ N 100º 29´ 15´´ E on the world map. Sungai Petani is the largest town of Kedah with population of 443,458 in 2010. Kulim is located at 5º 21´ 36´´ N 100º 32´ 59´´ E in the southwest of Kedah. On the west it borders the Penang. Baling is located at latitude 5º 40́ 0´´ N and longitude 100º 55´ 0´´ E and lies to the south-east of Kedah, approximately 56 km from Sungai Petani and close to the border

of Thailand. It has a total area of 1530 km^2 (590 Square miles) with population (2009) of 204,300. Figure 3.1 shows map of study area.

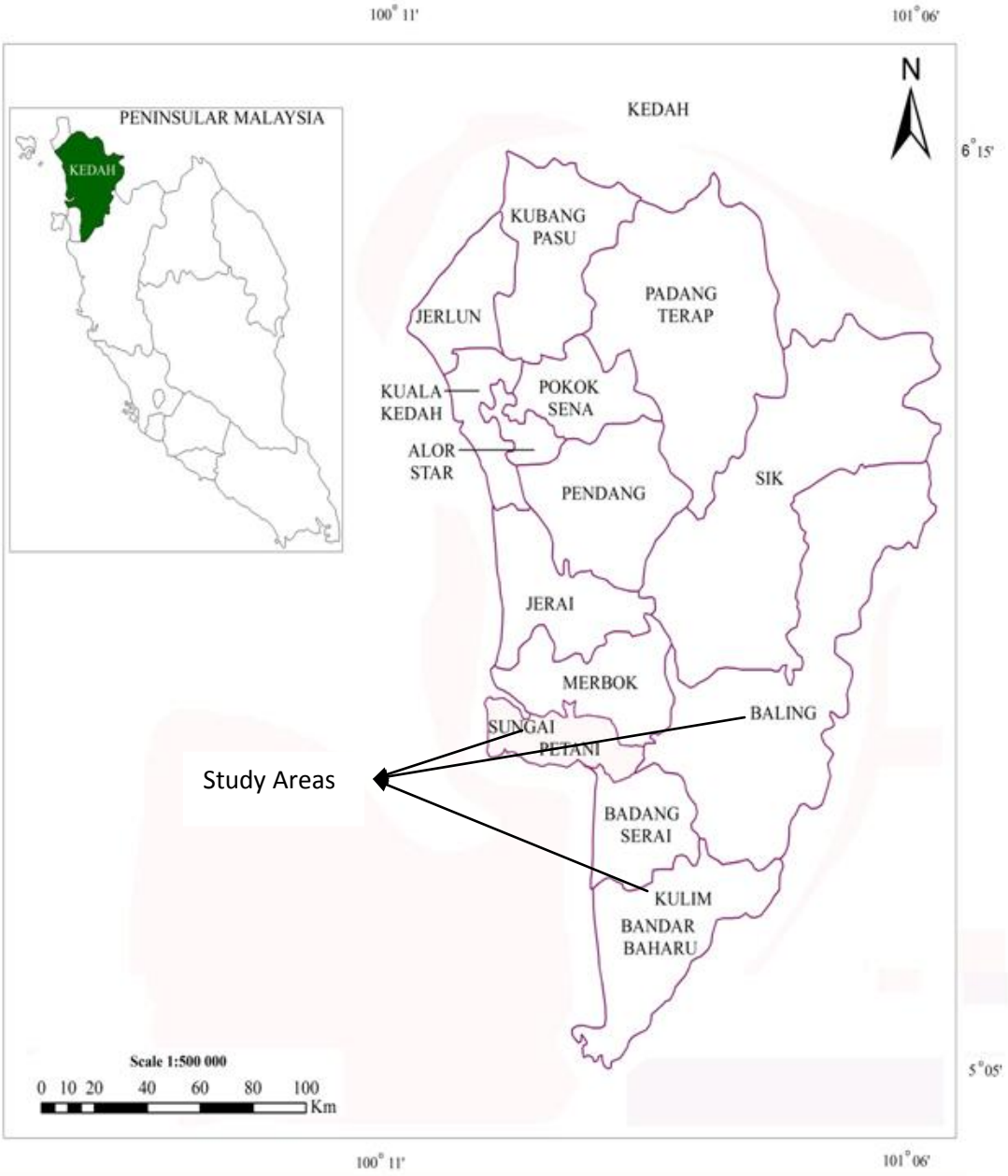

Figure 3.1: Map of Kedah, showing the study areas

3.2 Collection of Samples, Materials and Methods

3.2.1 Collection of Soil Samples

A total of 73 soil samples for the measurement of radon concentration, natural radioactivity and heavy metals were collected from uncultivated and cultivated (Chili, Banana and palm oil forms) areas of Sungai Petani, Kulim and Baling. Soil samples collected for the measurement of heavy metals were stored in insulated ice cooler in order to protect it from sun heat and brought to the Medical Physics Laboratory on the same day and stored at 4 ºC until processing, dried at 110 °C for 2 hr and passed through sieve of size 0.249 mm after grinding (Jan et al., 2010). Each sample was weighted by using electrical balances. The geographic sites of the soil sampling sites are tabulated in Table 3.1.

Table 3.1: Geographic sites of soil sampling locations

S No	Site Name	Sample Code	Co-ordinates
	Uncultivated soil		
1	Industrial Area Sungai Petani (5 samples)	SPI	N 05° 36' 33.2", E 100° 30' 12.5" N 05° 36' 22.3", E 100° 30' 10.8" N 05° 35' 32.1", E 100° 30' 09.9" N 05° 36' 05.2", E 100° 29' 48.1" N 05° 35' 09.5", E 100° 27' 13.6"
2	Kampung Kilang Makau, Sungai Patani	SP11	N 05° 35' 19.4", E 100° 29' 02.7"
3	Kampung Kubang Sapi, Sungai Patani	SP10	N 05° 33' 51.3", E 100° 33' 13.2"
4	Kampung Bakar Kapor, Sungai Patani	SP7	N 05° 38' 25.7", E 100° 28' 50.4"
5	Kampung Pantai Cicak, Sungai Patani	SP9	N 05° 36' 36.5", E 100° 37' 19.5"
6	Taman Seri Baiduri, Sungai Patani	SP15	N 05° 37' 11", E 100° 37' 19.5"
7	Taman Sinar Permata, Sungai Patani	SP13	N 05° 36' 02.2", 100° 28' 09.9"
8	Kumpung Tanah Licin, Sungai Patani	SP8	N 05° 35' 57.2", E 100° 36' 29.5"
9	Kolej Komuniti Baling	B1	N 05° 39' 18.4", E 100° 52" 25.7"